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Crude Oils of Domanik Genotype from Middle Devonian Deposits of the Timan–Pechora Oil-and-Gas Province

D. A. Bushnev[a],* and O. V. Valyaeva[a],**

[a] *Institute of Geology of Komi Science Centre of the Ural Branch of the Russian Academy of Sciences, Komi Republic, Syktyvkar, 167982 Russia*
**e-mail: boushnev@geo.komisc.ru; **e-mail: valyaeva@geo.komisc.ru*

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Abstract—The composition of biomarker hydrocarbons in crude oil samples from Middle Devonian deposits of the Western Tebuk, Dzh”er, and Michayu oil fields of the Omra–Lyzha saddle in the Timan–Pechora oil-and-gas province was studied. Significant effect of the Upper Devonian Domanik source rocks on the formation of the composition of these crude oils was revealed. The main parameters demonstrating the genetic relationship of the oils from Middle Devonian reservoirs with this source rocks are distribution of steranes $C_{27} : C_{28} : C_{29}$, ratio of isoprenoid to normal alkanes, and distribution of normal alkanes. All these characteristics sharply distinguish the crude oils under consideration from paraffinous oils syngenetic to Middle Devonian deposits, characteristic of deposits in the southern part of the Pechora–Kozhva megarampart.

Keywords: Timan–Pechora oil-and-gas province, biomarker hydrocarbons, carbon isotope composition

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The biomarker hydrocarbon composition and other geochemical features of crude oils and source rocks from various oil-and-gas complexes (OGCs) of the Timan–Pechora oil-and-gas province (TPP) were studied in detail previously [1–4]. The previously studied crude oils from the Middle Devonian–Lower Frasnian terrigenous complex of the southern part of the Pechora–Kozhva megarampart have extremely high content of paraffin hydrocarbons (HCs), and the oils themselves are of light color with relatively high congealing points and low resin content. They are also characterized by very low content of isoprenanes and a high-molecular-mass normal alkane “hump” in the chromatograms [5]. On the other hand, crude oils from the Domanik–Tournaisian complex, demonstrating genetic relationship with the Middle Frasnian Domanik organic matter, exhibit properties essentially differing from those of Middle Devonian paraffinous oils [6].

When studying crude oils from the Middle Devonian–Lower Frasnian terrigenous oil-and-gas complex of the TPP Omra–Lyzha saddle, we revealed interesting features of some of them. Here we make an attempt to interpret these features, based on a detailed study of the hydrocarbon and isotope composition of crude oils from the Western Tebuk, Dzh”er, and Michayu oil fields in comparison with other crude oils of the terrigenous Middle Devonian–Lower Frasnian complex and with crude oils typical of the overlying Domanik–Tournaisian complex of deposits [7].

EXPERIMENTAL

Investigation objects. As investigation objects we chose crude oils from Middle Devonian deposits of the Western Tebuk and Dzh”er oil fields of the Tebuk step and of the Michayu oil field of the Michayu–Pashnya rampart. These crude oils are light and medium (density 0.847 and 0.855 g/cm^3, respectively) with medium sulfur content (0.61–0.74%) (Table 1).

From the territorial standpoint, the Western Tebuk and Dzh”er oil fields discovered in the 1960s are associated with the Tebuk step, and the Michayu oil field, with the Michayu–Pashnya rampart (Fig. 1). The Tebuk step and Michayu–Pashnya rampart are second-order tectonic elements of the Omra–Lyzha saddle of the Izhma–Pechora syneclise located in the western part of TPP [8]. The

Table 1. Group composition and geochemical parameters of crude oils from deposits of the Middle Devonian–Lower Frasnian terrigenous complex of TPP (GC and GC-MS data)

Parameter[a]	Western Tebuk (D_2ef)	Dzh"er (D_2ef)	Michayu (D_2st)
Group composition of crude oils			
Density, g/cm^3	0.847	0.855	0.855
Sulfur content, %	0.69	0.61	0.74
Oils, %	67.4	63.2	65.9
Resins, %	18.8	17.6	13.7
Asphaltenes, %	2.1	1.4	2.9
Saturated HCs, %	35.0	36.7	41.1
Aromatic HCs, %	24.5	25.7	24.0
n-Alkanes and isoprenanes			
C_{12}–C_{18}	47	48	45
C_{19}–C_{24}	35	33	37
C_{25}–C_{32}	18	19	18
n-Alkanes/isoprenanes	4.1	4.2	3.1
Pr/Ph	1.00	1.18	1.11
(Pr + Ph)/(C_{17} + C_{18})	0.78	0.74	1.09
Pr/C_{17}	0.75	0.76	1.08
Ph/C_{18}	1.22	1.40	0.92
$K_{odd}C_{17}$	1.03	1.05	1.09
$K_{odd}C_{29}$	1.12	1.12	1.13
CPI	1.08	1.06	1.06
Steranes and hopanes			
αββ steranes C_{27}:C_{28}:C_{29}	32 :18:50	33:18:49	33:18:49
Dia/reg	0.47	0.45	0.55
Steranes/hopanes	0.25	0.23	0.24
C_{29}/C_{30} hopane	0.69	0.69	0.65
Tri-/pentacyclanes	0.22	0.23	0.19
βα C_{30}, %	7.71	8.04	8.77
Ts/Tm	0.70	0.86	0.73
αββ/(αββ+ααα) C_{29}	0.54	0.57	0.57
ααα C_{29} 20S/(20S+R)	0.50	0.44	0.50
22S/(22S + 22R) C_{31} hopane	0.55	0.56	0.56

[a] Isoprenanes = *iso*-C_{15} + *iso*-C_{16} + *iso*-C_{18} + *iso*-C_{19} + *iso*-C_{20}; Pr, pristane; Ph, phytane; $K_{odd}C_{17} = 2C_{17}/(C_{16} + C_{18})$; $K_{odd}C_{29} = 2C_{29}/(C_{28} + C_{30})$; CPI = $0.5(\Sigma C_{25, 27, 29, 31}/\Sigma C_{24, 26, 28, 30} + \Sigma C_{25, 27, 29, 31}/\Sigma C_{26, 28, 30, 32})$; Dia/Reg, ratio of diasteranes C_{27} to regular steranes C_{27}; Ts, 18α(H)-22,29,30-trisnormethylhopane; Tm, 17α(H)-22,29,30-trisnormethylhopane.

Omra–Lyzha saddle occupies the central, eastern, and southern parts of the Izhma–Pechora syneclise. It is a large (70 × 500 km) first-order positive structure elongated in the meridional direction, separating the Upper Pechora depression from the Neritsa, Izhma, and Kipievo steps of the Izhma–Pechora syneclise. The northern boundary of the saddle is the Pechora–Kozhva megarampart. From the tectonic standpoint, the Michayu rampart is associated with the eastern side of the Izhma–Pechora syneclise [9]. The rampart is complicated by local uplifts stretching from NNW to SSE. The total rampart length is about 100 km. As seen on the geological section along the 22-RS profile (Fig. 1), deposits D_2 of the oil field group under consideration lie gypsometrically considerably higher (by up to 1000 m) than D_3fr-fm deposits of the cis-Urals edge deflection, which can affect the composition of HC fluids in the deposits.

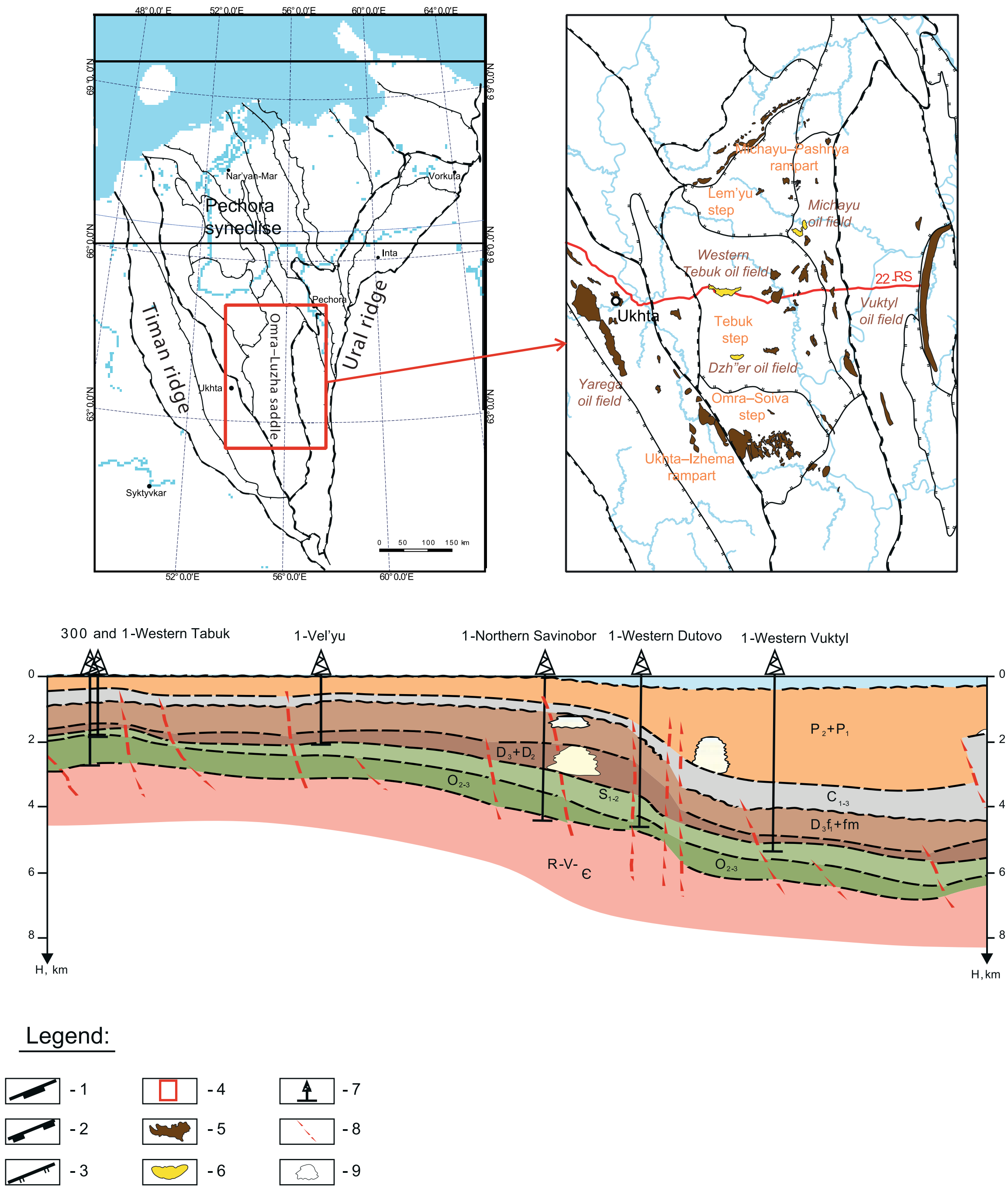

Fig. 1. Scheme of location of investigation objects and geological section along the line of the 22-RS profile. Designations: (1) boundaries of first-order tectonic elements, (2) boundaries of second-order tectonic elements, (3) boundaries of third-order tectonic elements, (4) region of works, (5) oil fields, (6) examined fields, (7) wells, (8) tectonic disturbances, and (9) reefs. Geological section along the line of the 22-RS profile [10].

Oil fractionation. From a weighed portion of crude oil, we precipitated asphaltenes with a 40-fold volume of n-hexane. Then, the resulting maltene fraction was separated on a column packed with alumina into nonpolar (oils, 50 mL of a 20% solution of dichloromethane in n-hexane) and polar (resins, 50 mL of a 1 : 1 ethanol–benzene mixture) fractions. The nonpolar fraction was then separated on a column packed with silica gel into saturated (eluent n-hexane) and aromatic (eluent benzene) HCs.

Analysis. The saturated HC fraction was analyzed by gas chromatography (GC) and gas chromatography–mass spectrometry (GC-MS).

Gas-chromatographic analysis was performed with a Crystal-2000M device under the following conditions: DB-5 column, 30 m × 0.32 mm, stationary phase film thickness 0.25 μm, programmed heating from 110 to 300°C at a rate of 5°C/min, injector and detector temperature 300°C. GC-MS analysis was performed with a Shimadzu 2010 Ultra device under the following conditions: HP-5 column, 30 m × 0.25 mm, stationary phase film thickness 0.25 μm, programmed heating from 110 to 300°C at a rate of 5°C/min, injector temperature 300°C, detector temperature 250°C. Sterane HCs were detected using the mass chromatograms at *m/z* 217 and 218, and terpane HCs, using the mass chromatograms at *m/z* 191.

The carbon isotope composition (CIC) of oil fractions was measured in a continuous helium flow (CF-IRMS) using an analytical complex consisting of a Flash EA 1112 elemental analyzer connected with a Delta Y Advantage mass spectrometer (Thermo Fisher Scientific) via a Conflo IV gas switch. In our study, we used USGS-4O international standard (L-glutamic acid) and acetanilide laboratory standard (C_3H_9NO). The $\delta^{13}C$ values are given in ‰ relative to V-PDB standard. The measurement error is ±0.15‰ (1σ). Operator I.V. Smoleva.

RESULTS AND DISCUSSION

Group Composition

The major components of the crudes are oils (Table 1). The asphaltene content is low (from 1.4 to 2.9%). The resin content reaches 18.8%. The percentage of the saturated fraction in the crudes studied is in the range 35.0–41.1%, and the percentage of the aromatic fraction, in the range 24.0–25.7%.

The group composition of paraffinous crude oils from the southern part of the Pechora–Kozhva megarampart [5] sharply differs from that of the samples from Middle Devonian deposits examined in this study. The previously studied paraffinous crude oils from the Yugid and Kyrtael' oil fields contain higher amounts of saturated HCs, considerably lower amounts of resins and aromatic HCs, and lower amounts of asphaltenes. Data obtained for numerous previously studied crude oils from Upper Devonian deposits, belonging to Domanik genotype, show that these oils also essentially differ from paraffinous Middle Devonian oils in that they contain higher amounts of asphaltenes, resins, and aromatic HCs [6, 7].

The crude oils studied in this work are similar in the group composition specifically to crude oils of Domanik genotype.

Distribution of n-Alkanes and Isoalkanes

According to the GC data, the crude oils are characterized by the distribution of n-alkanes with the prevalence of low-molecular-mass compounds in the region of C_{12}–C_{20} with maxima at *n*-C_{15} and *n*-C_{17} (Fig. 2). However, there is no appreciable prevalence of odd HCs (Table 1). The oddness coefficient $K_{odd}C_{17}$ is close to 1, which suggests insignificant contribution of algal organic matter to the initial biomass [11, 12].

The concentration of high-molecular-mass normal alkanes C_{25}–C_{32} is insignificant and varies from 18 to 19% (of the sum of n-alkanes).

According to previous assumptions [5, 13, 14], TPP paraffinous oils from deposits of the Middle Devonian–Lower Frasnian terrigenous oil-and-gas complex are genetically related to the OM of the surrounding terrigenous deposits, which is manifested in the distribution of n-alkanes and isoprenanes. However, the alkane distribution that we observed in crude oils from the Western Tebuk, Dzh"er, and Michayu oil fields is characteristic of Domanik-type crude oils [6, 7] (Fig. 2).

Domanik deposits (D_3dm, formally corresponding to deposits of the Semiluki horizon of the Middle Frasnian stage) widely occur in the Timan–Pechora sedimentation basin. The Domanik horizon in TPP was studied from the geochemical standpoint in numerous works [7, 15–22, etc.]. From the lithological standpoint, the Domanik horizon is mainly constituted by argillites (mainly siliceous), siliceous marls, clayey limestones, and oil

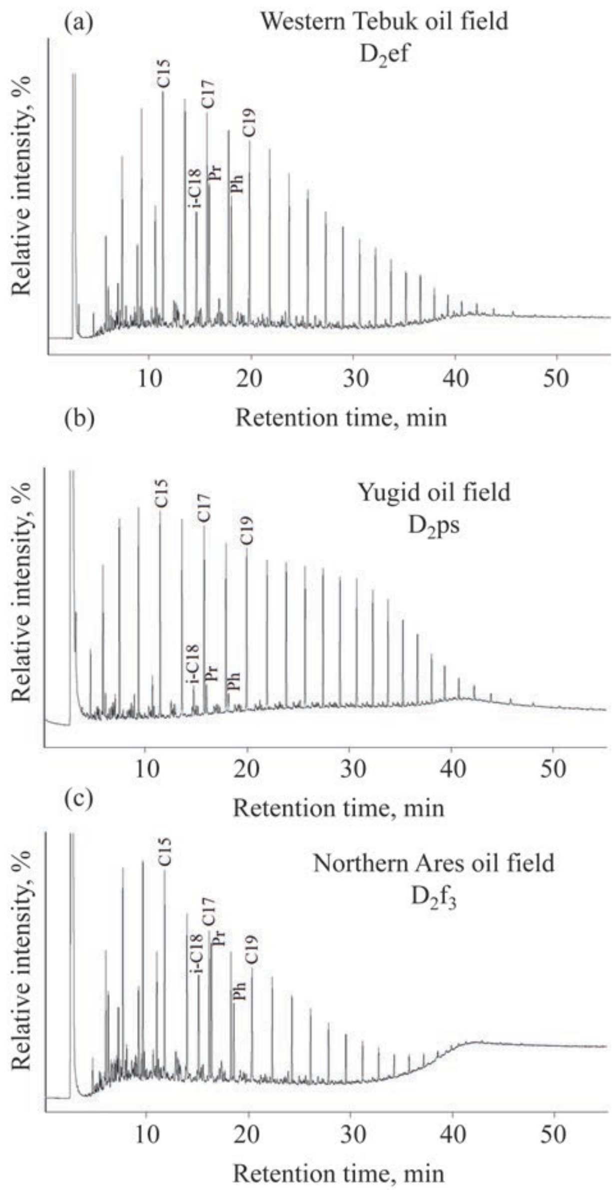

Fig. 2. Distribution of n-alkanes and isoalkanes in the saturated fraction of crude oils. C (number), n-alkaines; Pr, pristane; Ph, phytane; iso-C (number), isoalkanes. (a) Western Tebuk, (b) Yugid, and (c) Northern Ares oil fields.

shales [13]. The rocks are characterized by increased OM content and different stages of catagenesis.

According to [23], in the Izhma–Pechora generation source the generating strata are developed in two OGCs: Middle Devonian–Lower Frasnian terrigenous and Domanik–Tournaisian carbonate complexes. The OM type in both complexes is mixed, with the prevalence of the sapropel material. The crude oils revealed in the Western Tebuk, Michayu, and Dzh”er oil fields, according to [23], are syngenetic to the surrounding rocks. The occurrence of lateral migration in the Domanik–Tournaisian OGC is noted; it is traced almost throughout the depression territory, except its northern part [23].

Bushnev [15] believes that crude oils from the Izhma–Pechora depression of the terrigenous OGC D_2-D_3f_1 are syngenetic to their complex, with mixed OM (humites–sapropelites) serving as an HC source.

As reported in [24–26], the oil and gas deposits in the south of TPP were formed in a number of stages by lateral and vertical migration of HCs both from their own sources (region of the Omra–Soiva step) and from remote regions (long-range migration) of the south of the Izhma–Pechora syneclise and Upper Pechora depression. This caused the formation of mixed HCs of different phase state and catagenetic maturity in the deposits.

Distribution of Polycyclic Biomarkers

The distribution of αββ steranes of the composition C_{27}–C_{29} is similar (Table 1). Ethylcholestane (C_{29}) prevails over adjacent homologs; its relative content in crude oils reaches 50%. Such distribution is characteristic of Domanik-type crude oils [6, 7]. As in crude oils from Tatarstan [27], in the oils under consideration the concentrations of C_{28} sterane are relatively low. The other parameters traditionally classed with so-called genetic parameters, such as the ratio of diasteranes to regular steranes, steranes/hopanes ratio, and ratio of tricyclic to pentacyclic terpanes, are quite typical of Domanik-genotype crude oils [6, 7].

Terpenoids consist of tricyclic and pentacyclic hopanoids (Fig. 3). In the mass chromatograms recorded at *m/z* 191, we identified tricycloalkane HCs C_{19}–C_{25} and hopanes from C_{27} to C_{35}. For all the crude oils studied, hopane αβ C_{30} prevails.

Parameters calculated from data on the distribution of stereoisomers of sterane and terpane HCs are used in the geochemical practice to determine the degree of maturity of crude oils and initial OM [28, 29]. As judged from the ratios αββ/(αββ + ααα) C_{29}, ααα C_{29} 20S/(20S + 20R), 22S/(22S + 22R) C_{31} hopane, and Ts/Tm, the crude oils from the Middle Devonian reservoirs do not significantly differ in the thermal maturity from the Domanik-genotype crude oils from Upper Devonian reservoirs, and their formation is associated with the kerogen transformation in the beginning and middle of the main phase of the oil formation, as for Domanik crude oils.

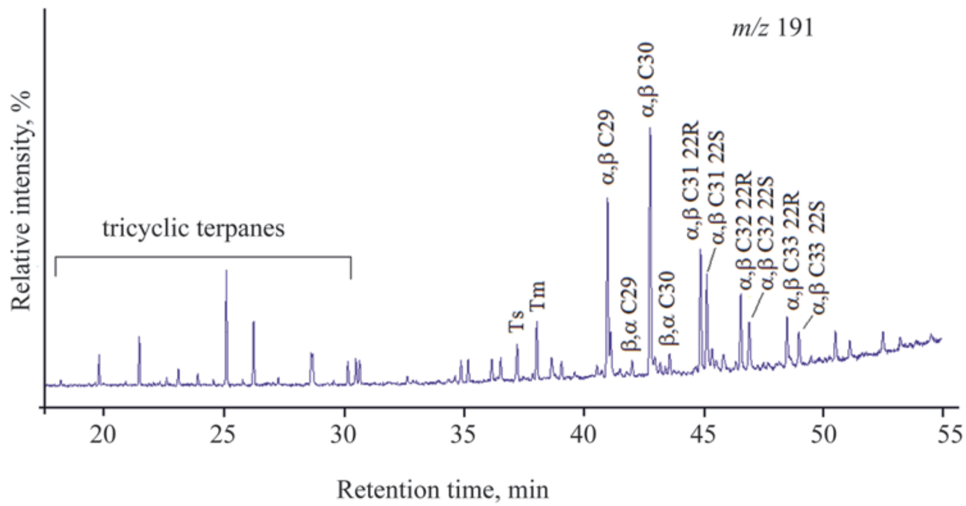

Fig. 3. Mass chromatogram recorded at *m/z* 191, demonstrating typical distribution of terpenoids in Domanik-type crude oils.

Isotope Composition of Carbon in Oil Fractions

The distribution of carbon isotopes in fractions of crude oils was studied. Figure 4 shows the CIC of fractions in a series of Domanik-genotype crude oils from Upper Devonian reservoirs [6, 7], in paraffinous crude oils from Middle Devonian–Lower Frasnian OGC deposits, which are probably genetically associated with OM of terrigenous deposits [5], and also in Domanik-type crude oils from Upper Devonian reservoirs that we studied. As can be seen, carbon of the aliphatic and resin fractions of the oils (Fig. 1) has intermediate CIC between those of paraffinous oils and TPP Domanik oils. The asphaltene carbon in the oils under consideration is close to that in Domanik oils. Presumably, the isotope data presented suggest mixing of Domanik-genotype oil and certain amounts of high-paraffin asphaltene-free differences syngenetic to the surrounding deposits in the reservoirs. However, it seems incorrect to speak of specific proportions of fluid mixing, based on the data presented.

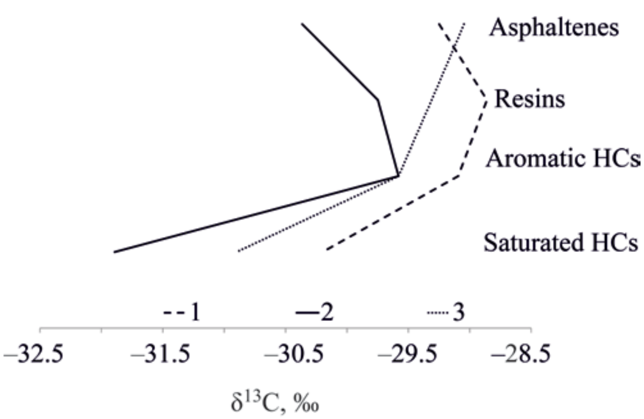

Fig. 4. Distribution of carbon isotopes in oil fractions: (1) mean data for Domanik-genotype oils [6], (2) mean data for paraffinous oils from Middle Devonian deposits [5], and (3) mean data for oils from Table 1.

CONCLUISIONS

The group composition, biomarker hydrocarbon composition, and carbon isotope composition were studied for fractions of crude oils from a series of Middle Devonian reservoirs of the Omra–Lyzha saddle of the Timan–Pechora province. For comparison with the oils studied, paraffinous oils from Middle Devonian reservoirs and Domanik-genotype Upper Devonian oils were chosen. This choice was governed by the available data on possible generation of hydrocarbons in these two main complexes on the territory under consideration.

Analysis of the whole set of data on the composition of biomarker hydrocarbons shows that crude oils from the Omra–Lyzha saddle, located in Middle Devonian reservoirs, are characterized by a set of genetic parameters (distribution of C_{27} : C_{28} : C_{29} steranes, ratios dia-/regular steranes, steranes/hopanes, tri-/pentacyclanes) virtually equal to the mean values of these parameters for Domanik-genotype crude oils of the Timan–Pechora province. The crude oils under consideration are also similar to Domanik oils in the group composition but differ essentially from paraffinous oils syngenetic to the Middle Devonian–Lower Frasnian terrigenous complex. The whole set of this data suggests the decisive contribution of the organic matter of Domanik deposits to the oil-bearing capacity of the Middle Devonian in the Omra–Lyzha saddle and hence allows speaking of the geochemically proved migration into underlying oil-and-gas complexes. The contribution of the organic matter of Middle Devonian deposits, syngenetic to the complex, is also proved by the isotope analysis of carbon in crude oils. The contribution of the Domanik organic matter to the accumulation of crude oils in Middle Devonian reservoirs can be attributed to the flow of HC fluids from D_3fr-fm deposits of the cis-Urals edge deflection to traps of the Middle Devonian age.

AUTHOR INFORMATION

D.A. Bushnev, ORCID: https://orcid.org/0000-0002-3860-944X

O.V. Valyaeva, ORCID: https://orcid.org/0000-0003-3479-4673

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CONFLICT OF INTEREST

The authors declare no conflict of interest requiring disclosure in this article.

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